

# Nearly-critical spin and charge fluctuations in $\text{KFe}_2\text{As}_2$ observed by high-pressure NMR

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We report a high-pressure  $^{75}\text{As}$  NMR study on the heavily hole-doped iron pnictide superconductor  $\text{KFe}_2\text{As}_2$  ( $T_c \approx 3.8$  K). The low-energy spin fluctuations are found to decrease with applied pressure up to 2 GPa, but then increase again, changing in lockstep with the pressure-induced evolution of  $T_c$ . Their diverging nature suggests close proximity to a magnetic quantum critical point at a negative pressure of  $P \simeq -0.6$  GPa. Above 2.4 GPa, the  $^{75}\text{As}$  satellite spectra split below 40 K, indicating a breaking of As site symmetry and an incipient charge order. These pressure-controlled phenomena demonstrate the presence of nearly-critical fluctuations in both spin and charge, providing essential input for the origin of superconductivity.

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The presence of multiple active orbitals in iron-based superconductors (FeSCs) [1–4] causes a range of highly non-trivial characteristics. The combination of Fermi-surface nesting, Hund coupling, and Coulomb interactions produces a rich variety of electronic and magnetic properties [5] and a complex interplay among the lattice structure, magnetism, and superconductivity [6, 7].

Heavily doped FeSCs, whose Fermi surfaces are quite different from optimally doped materials, challenge the existing understanding.  $\text{KFe}_2\text{As}_2$  has large hole doping (0.5 hole/Fe), but far from being a regular metal it shows heavy-fermion characteristics below a low coherence temperature of order 60 K [8] and superconductivity at a low but finite  $T_c$  of 3.8 K [9, 10]. The absence of electron pockets around  $(\pi, \pi)$  [11] suggests that spin fluctuations from interband nesting are unlikely, but low-energy electronic correlations are surprisingly strong. Similarly strong low-energy spin fluctuations [12, 13] and significant electron mass enhancement (average  $m^*/m_e \sim 12$ ) [14, 15] have been reported in  $\text{KFe}_2\text{As}_2$ , and appear to strengthen with  $x$  in  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  [15]. Controversy over  $s^\pm$ -wave (favored by interband scattering) or  $d$ -wave pairing symmetry [8, 16–18] raises questions about the effects of electron correlations on superconductivity.

Recent high-pressure studies of  $\text{KFe}_2\text{As}_2$  discovered an anomalous reversal of  $T_c$ , which has a minimum at 1.8 GPa [18]. Scenarios proposed to explain this include a change of pairing symmetry [18, 19] and a  $k_z$  modulation of the superconducting gap [20]. Although spin fluctuations are essential to FeSC superconductivity, no measurements under pressure have yet been reported.

In this Letter, we present a high-pressure study of

$\text{KFe}_2\text{As}_2$  by nuclear magnetic resonance (NMR). The  $^{75}\text{As}$  spectra and spin-lattice relaxation rate ( $1/T_1T$ ) are measured under pressures up to 2.42 GPa, revealing three surprising features. First,  $1/T_1T$  is dominated by strong low-energy spin fluctuations, suggesting incipient antiferromagnetic order at a quantum critical point near  $-0.6$  GPa. Second, the spin fluctuations show exactly the same reversal behavior as  $T_c$ , and indeed identical evolution at all pressures. Third, a line splitting of the  $^{75}\text{As}$  satellite spectra below 40 K for pressures above 2.4 GPa indicates a breaking of four-fold symmetry. This effect is caused by charge order, whose fluctuations we propose are strong around the  $d^{5.5}$  electron filling of  $\text{KFe}_2\text{As}_2$ . This emergent charge order is accompanied by the enhancement of spin fluctuations and hence of  $T_c$ , demonstrating the importance of nearly-critical charge fluctuations in heavily hole-doped FeSCs.

Our  $\text{KFe}_2\text{As}_2$  single crystals were synthesized by the self-flux method [21]. We measure very large residual resistivity ratios of 1390, indicating extremely high sample quality. We performed high-pressure NMR measurements using a NiCrAl clamp cell, which reaches a maximum pressure of 2.42 GPa at  $T = 2$  K; to obtain a maximally hydrostatic pressure, we chose Daphne oil as the medium. The actual pressure was calculated from the  $^{63}\text{Cu}$  nuclear quadrupole resonance (NQR) frequency of  $\text{Cu}_2\text{O}$  powders [22].  $P(T)$  changes negligibly below 150 K and here we use the pressure values measured at  $T = 2$  K. The superconducting transition temperature,  $T_c$ , was determined from the RF inductance of the NMR coil. The  $^{75}\text{As}$  NMR and NQR signals were measured by the spin-echo technique. All NMR data were taken with a mag-

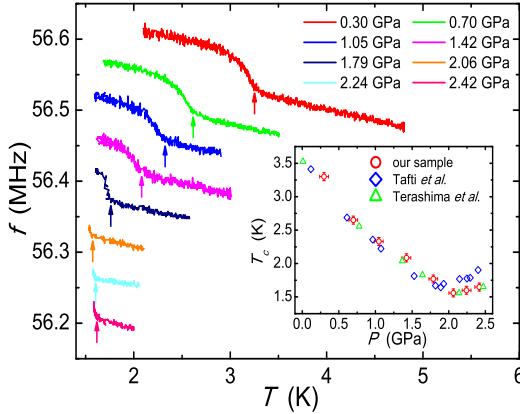


FIG. 1. (color online) Resonance frequency  $f$  of the detuned NMR coil at different pressures, as a function of temperature, measured at zero field. Lines are offset vertically for clarity and arrows indicate  $T_c$ . Inset: measured  $T_c$  as a function of pressure (open circles), compared with  $T_c$  data reported from transport measurements [18] (open diamonds) and from  $ac$  susceptibility measurements [19] (open triangles).

magnetic field of 10.6 T, which is well beyond  $H_{c2}$ , applied in the crystalline  $ab$ -plane. The spin-lattice relaxation rate  $1/T_1$  was measured by the spin-inversion method, and the spin recovery is fitted well by a single  $T_1$  component at all pressures, indicating high sample homogeneity.

$T_c$  data for KFe<sub>2</sub>As<sub>2</sub> under pressure are determined from the resonance frequency  $f$  of the detuned NMR circuit on cooling. The superconducting transition causes a sharp increase of  $f$  [Fig. 1] due to the decrease of coil inductance.  $T_c$  as a function of pressure (inset, Fig. 1) has an initial decrease, at a rate  $dT_c/dP \simeq -1.6$  K/GPa, but reaches a minimum of  $T_c = 1.5 \pm 0.05$  K at  $P \simeq 2.1$  GPa, then increases slowly ( $dT_c/dP \simeq 0.3$  K/GPa). Our data for  $T_c(P)$  are fully consistent with  $ac$  susceptibility measurements [19], whereas transport measurements [18] found a lower reversal pressure of  $P \approx 1.6$  GPa and a larger  $T_c$  beyond this. We suggest that our results and those of Ref. [19] represent more closely the bulk  $T_c(P)$ , whereas transport measurements are more susceptible to boundary superconducting phases in multi-domain samples and to non-hydrostatic pressure conditions [23].

Figure 2(a) shows  $1/T_1T$  for a range of pressure values. Qualitatively, it rises very strongly on cooling, suggesting close proximity to a magnetic ordering transition. Quantitatively, we obtain an excellent fit to the low-temperature data ( $T \leq 40$  K) with the function  $1/T_1T = a + b/(T - \theta)$  [inset, Fig. 2(a)], where  $a$  is a temperature-independent contribution characteristic of itinerant electrons and the  $b$  term is a Curie-Weiss function describing contributions from spin fluctuations [24]. This behavior contrasts starkly with the weak temperature-dependence of the Knight shift,  ${}^{75}K$  [shown in Fig. 3(d)]. Because  ${}^{75}K$  measures the suscepti-

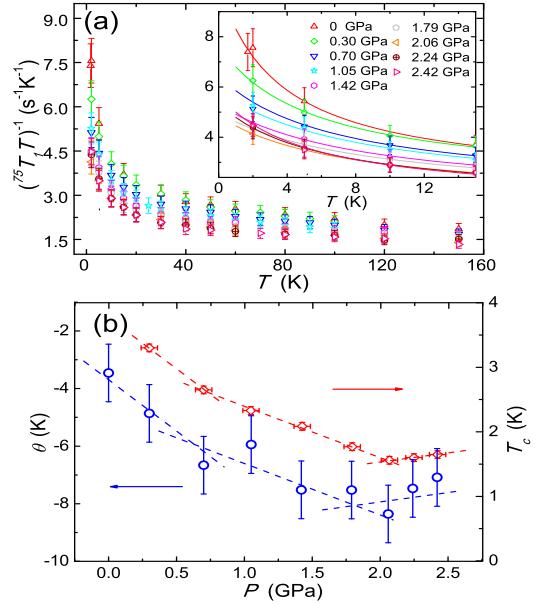


FIG. 2. (color online) (a) Normal-state  $1/T_1T$  as a function of temperature from 2 K to 150 K, measured at different pressures. Inset: zoom of low-temperature data. Solid lines are fits with the Curie-Weiss form  $1/T_1T = a + b/(T - \theta)$ . (b) Values of  $\theta$  extracted from  $1/T_1T$  (left axis) and of the measured  $T_c$  (right axis) as functions of pressure.

bility at  $q = 0$ , whereas  $1/T_1T$  has contributions from all  $\mathbf{q}$ , Fig. 2(a) indicates that the spin fluctuations are almost exclusively antiferromagnetic [25, 26].

To describe the evolution of  $1/T_1T$  with increasing pressure, we find that  $a$  decreases only slightly, suggesting a small reduction in density of states on the Fermi surface. Changes of  $b$  are very weak, and hence the low-energy spin fluctuations are characterized almost exclusively by the Curie-Weiss temperature  $\theta$ . The extracted value of  $\theta$  is small and negative at all pressures [Fig. 2(b)], with an initial rapid decrease from  $-3.5$  K, at  $-5.3$  K/GPa, whose rate slows until a minimum is reached at 2 GPa; here  $\theta$  reverses and increases slowly (1 K/GPa), i.e. spin fluctuations are suppressed by low pressures but are enhanced again at high pressures [Fig. 2(a)].

Figure 2(b) contains two key messages. First, from the initial value of  $d\theta/dP \simeq -5.3$  K/GPa, one may estimate that a magnetic quantum phase transition occurs at negative pressure ( $P_c = -0.6$  GPa). Such a low  $P_c$  implies that magnetic order could be achieved by chemical pressure. The small value of  $\theta$  at ambient pressure reflects that  $1/T_1T$  is dominated by nearly-critical magnetic fluctuations, which is a major surprise in a system as heavily hole-doped as KFe<sub>2</sub>As<sub>2</sub>. The initial decrease of  $\theta$  under pressure is consistent with cyclotron resonance measurements, which found a correspondingly reduced mass enhancement [19]. CsFe<sub>2</sub>As<sub>2</sub> is a material with a lower chemical pressure than KFe<sub>2</sub>As<sub>2</sub> (the larger Cs ions expand the in-plane lattice parameters) and shows

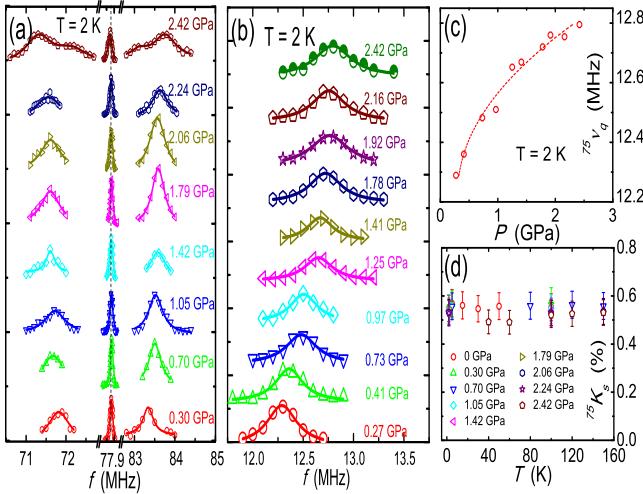


FIG. 3. (color online) (a)  $^{75}\text{As}$  spectra measured at different pressures under a field of 10.6 T applied in the  $ab$ -plane and at  $T = 2$  K. Data for different pressures are offset for clarity. (b)  $^{75}\text{As}$  NQR spectra measured at different pressures in zero field and at  $T = 2$  K (data again offset). (c)  $^{75}\nu_q$  as a function of pressure. (d)  $^{75}\text{As}$  Knight shift as a function of temperature.

a larger mass enhancement [27], in agreement with our deduction, but no magnetic order. Although  $T_c(P)$  is qualitatively identical to that in  $\text{KFe}_2\text{As}_2$  [28], the lower  $T_c$  values and lower reversal pressure are not understood. The second key message is that  $\theta$  and  $T_c$  change in lock-step as functions of pressure; such behavior has been observed in other FeSCs [29] and has the unambiguous interpretation that magnetic fluctuations are the primary contributor to superconducting pairing.

Next we consider the  $^{75}\text{As}$  NMR spectra under pressure. For a nuclear spin of  $I = 3/2$ , each spectrum [Fig. 3(a)] consists of one center line, whose width is controlled by second-order corrections from electric-field gradients (EFGs), and two broad satellites subject to first-order corrections. At  $T = 2$  K, increasing pressure causes the satellites to shift further from the center line, a trend confirmed by the quadrupole frequency,  $^{75}\nu_q$ , which is determined by NQR at zero field [Fig. 3(b)].  $^{75}\nu_q$  increases monotonically with pressure [Fig. 3(c)], but at a decreasing rate. Because  $^{75}\nu_q$  measures the local EFG at the  $^{75}\text{As}$  site, this result is consistent with the gradual decrease of the in-plane lattice parameter under pressure reported by x-ray measurements [28]. The Knight shift, deduced from the center lines, is almost completely constant for all pressures and temperatures [Fig. 3(d)], indicating that the Fermi surface and the  $q = 0$  spin fluctuations undergo minimal changes at these pressures.

Below 2.24 GPa, there is little change in line shapes [Fig. 3(a)]. At 2.42 GPa, however, each satellite broadens and splits unambiguously into two peaks with a separation of 1 MHz at  $T = 2$  K. By measuring the  $^{75}\text{As}$  satellite spectra at 2.42 GPa for different temperatures

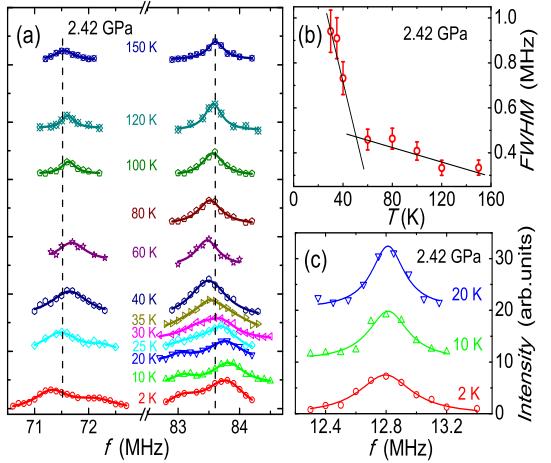


FIG. 4. (color online) (a)  $^{75}\text{As}$  NMR satellite spectra at the highest pressure  $P = 2.42$  GPa, measured at different temperatures. (b) FWHM of the  $^{75}\text{As}$  satellite (centered at 83.5 MHz) as a function of temperature. (c)  $^{75}\text{As}$  NQR spectra measured at low temperatures with  $P = 2.42$  GPa.

[Fig. 4(a)], we observe that each satellite is single-peaked at high  $T$  but splits into two peaks between 30 and 40 K. The full width at half maximum height (FWHM) of the 83.5 MHz satellite at 2.42 GPa, shown in Fig. 4(b), exhibits a pronounced kink below  $T \approx 40$  K, which marks the onset of splitting. Because Daphne oil solidifies above 150 K, a sharp line-splitting below 40 K can only be due to an intrinsic phase transition; simple strain effects or contributions from uniaxial pressure components could contribute to a line-broadening, but not to a splitting.

Such a broadening is indeed observed for the center line, whose FWHM increases from 80 kHz to 100 kHz; any splitting of this line could not exceed 20 kHz. The absence of measurable splitting in the center line, by comparison with the 1 MHz splitting of the satellites [Fig. 4(b)], leads us to conclude that the latter is a charge effect, rather than a magnetic effect. A double peak in the local EFG indicates two types of local charge distribution at different As sites, implying a charge ordering on NMR time scales. However, the  $^{75}\text{As}$  NQR spectra at zero field [Figs. 3(b) and 4(c)] remain single-peaked at low temperatures, demonstrating that the amplitude of the EFG is the same on all As sites. We deduce that the satellite splitting is caused by a broken in-plane (four-fold) symmetry of the EFG, creating two types of local charge environment with orthogonal orientations, which respond differently to the (fixed, in-plane) magnetic field.

Broken in-plane symmetry of the EFG should indicate a structural, orbital, or charge transition. In underdoped FeSCs,  $^{75}\text{As}$  satellite splitting with an in-plane field is observed just below the tetragonal to orthorhombic structural transition, where it is caused by twinning [30]. However, x-ray diffraction measurements find no evidence of a structural transition in  $\text{KFe}_2\text{As}_2$  in this pressure range

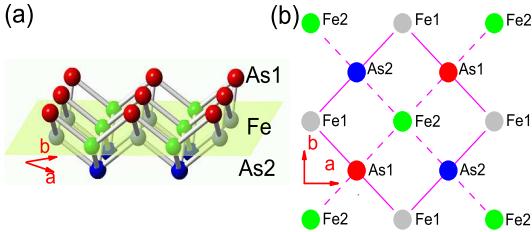


FIG. 5. (color online) (a) Schematic representation of the FeAs plane with checkerboard charge order on the Fe sites. (b) Planar projection showing charge-rich Fe1 and charge-poor Fe2 sites breaking four-fold symmetry at the As sites.

[18]. Thus charge ordering appears the most viable candidate to account for our observations.

For a qualitative understanding of our results, we note that  $\text{KFe}_2\text{As}_2$  has a hole doping concentration  $x = 0.5$ . Such half-doped configurations are generically less stable, and are often susceptible to charge-disproportionation fluctuations. These “mixed-valence” fluctuations are well known in manganites to stabilize the CE phase, which has a combined charge, orbital, and spin order [31], and a similar state has been proposed in a two-band FeSC model [32]. In  $\text{LiV}_2\text{O}_4$  ( $d^{1.5}$ ), valence fluctuations produce remarkable heavy-fermion behavior and highly anomalous electronic and magnetic properties [33–35]. We propose a checkerboard charge order on the Fe sites, shown in Fig. 5, where equally charge-rich and charge-poor sites alternate in two dimensions. The crystal structure of  $\text{KFe}_2\text{As}_2$  naturally contains two Fe sublattices with this pattern, because of the two inequivalent As sites, which in principle supports a checkerboard order. As a result, the four-fold symmetry of the charge density and thus of the EFG is broken on the As sites.

We stress that checkerboard charge order is the minimal scenario, as many different types of charge or orbital ordering with the compatible broken symmetry are possible. FeSCs are complex, multi-orbital systems where Hund coupling, on-site repulsion, Fermi-surface nesting, and orbital-selective correlation effects [36, 37] may all compete to induce order. Next-neighbor Coulomb interactions may be of particular importance for charge fluctuations in the pattern of Fig. 5. More generally, while checkerboard order provides an appealing real-space illustration of the mixed-valence phenomenon, in a multiband system it may take a rather different form with as few as one of the bands actually at commensurate filling.

Although checkerboard charge order breaks a local  $C_4$  rotational symmetry at the As sites (Fig. 5), it is quite different from the concept of nematicity in FeSCs, where anisotropic electronic properties are usually associated with an orthorhombic structural distortion [38]. The checkerboard pattern does not drive orthorhombicity, a result consistent with Ref. [28] and with the lack of anisotropy in any other static observables. Concerning

the nature of the charge-ordering transition, transport measurements find no additional charge localization in  $\text{KFe}_2\text{As}_2$  under pressure [20]. We suggest that the multiband system (transport dominated by itinerant bands) and the orbital-selective correlation effects mask any evidence of charge localization.

Regarding the time scale of charge-ordering, the symmetry-breaking at  $P = 2.42$  GPa appears static on the NMR time scale. However, we cannot exclude that this is only a quasi-static local order, which is pinned by a non-hydrostatic component in the applied pressure at its upper limit. As above, the charge order we observe in the satellite splitting has no other detectable experimental effects. Thus the question of static order, where the charge discrepancy  $\delta_i$  on each site satisfies  $\langle \delta_i \rangle \neq 0$ , or a quasi-static order with  $\langle \delta_i \rangle = 0$  but  $\langle \delta_i^2 \rangle \neq 0$ , remains open. In either case, charge ordering appears to enhance the spin fluctuations, setting in around 2.4 GPa while  $\theta$  increases above 2 GPa. Although no charge order is detected at ambient pressure, nearly-critical charge fluctuations ( $\langle \delta_i^2 \rangle \neq 0$ ) may exist over a wide pressure range in  $\text{KFe}_2\text{As}_2$  and contribute strongly to the nearly-critical spin fluctuations we observe.

These spin fluctuations are found [13] to be incommensurate, exhibiting peaks at  $(\pi(1 \pm \epsilon), 0)$  with  $\epsilon \simeq 1/6$ , so are quite different from the  $(\pi, 0)$  antiferromagnetic order of iron pnictides near  $x = 0$  [39]. Charge order in the  $d^{5.5}$  state should cause significant modification of local magnetic interactions and the incommensurability may result from stronger frustration on half of the next-neighbor bonds in Fig. 5. If electron filling is changed from  $d^6$  to  $d^5$ , the  $d_{xy}$  orbitals are thought to account for the observed increase in electronic correlations [14, 15]. When a multiband model is used to discuss magnetic order,  $d^6$  filling is found at the mean-field level to favor  $(\pi, 0)$  order whereas  $d^5$  favors a checkerboard  $[(\pi, \pi)]$  pattern [40].  $\text{KFe}_2\text{As}_2$  is close to neither filling and strong (nearly-critical) incommensurate fluctuations are entirely consistent with strongly competing states of different charge and spin order. We comment that a scenario of charge-ordering-induced magnetism may emerge from the correlation-driven scenario [36, 37] at  $x = 0.5$ .

Turning to the connection of charge and spin fluctuations to superconductivity, Fig. 2(b) shows that  $T_c(P)$  and  $\theta(P)$  have almost identical forms, and the pressures for minimum  $T_c$  and minimum spin fluctuations coincide within the error bar. This remarkable positive correlation is hard to interpret in any way other than spin fluctuations driving the change of  $T_c$  and hence a strong magnetic contribution to superconductivity. Our results suggest that charge fluctuations may enhance spin fluctuations, as the origin of the effects above 2 GPa, and thus are also constructive contributors to superconductivity.

A very similar simultaneous optimization of superconductivity and spin fluctuations was observed in  $\text{NaFe}_{0.96}\text{Co}_{0.06}\text{As}$  [29]. This electron-overdoped material

is close to good Fermi-surface nesting [41] and the spin fluctuations from itinerant electrons are thought to play the dominant role in modifying  $T_c$ . Because KFe<sub>2</sub>As<sub>2</sub> lacks electron pockets and viable nesting options, its spin fluctuations most likely originate in localized orbitals, which is fully consistent with weak pairing interactions and the low  $T_c$ . Valuable insight into the superconducting state could be obtained by ascertaining whether the pairing symmetry changes at the pressure-induced reversal in  $T_c$  [18–20], but we were unable to study this here.

Finally, we comment that the presence of strong charge and spin fluctuations is well known in unconventional superconductors, including many underdoped cuprates [42, 43], sodium cobaltates [44], and low-dimensional organic systems [45]. Thus it is not a complete surprise that a  $d^{5.5}$  material such as KFe<sub>2</sub>As<sub>2</sub> shows similar strong-correlation effects.

In summary, we have performed high-pressure <sup>75</sup>As NMR measurements on the heavily hole-doped iron superconductor KFe<sub>2</sub>As<sub>2</sub>. We find strong low-energy spin fluctuations indicating close proximity to a hidden magnetic ordering at low negative pressures. We find a charge order at high pressures, suggesting the predominance of nearly-critical charge (valence) fluctuations over much of the phase diagram. This is consistent with the low- $T$  heavy-fermion behavior, non-Fermi-liquid behavior at intermediate  $T$ , and low  $T_c$  value. We find that the spin fluctuations have a non-monotonic pressure dependence identical to that of  $T_c$ , reinforcing that pairing is of magnetic origin. We suggest that the half-integer filling ( $d^{5.5}$ ) causes nearly-critical charge fluctuations that are intrinsically linked to the nearly-critical spin state and hence to unconventional superconductivity.

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